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Recovery of Industrial and Recycled Al-Cu Alloys Subjected to Severe Plastic Deformation

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Abstract

Equal channel angular pressing (ECAP) is a well-known method to obtain high hardness levels through a strong refinement of grains. To obtain fine grains, a subsequent heating is performed after deformation. The main difficulty is to retain a sufficiently small grain size. Alloy purity is an important parameter in recrystallization kinetics. In the present work, an industrial and a recycled Al-4%Cu alloys were subjected to ECAP. The evolution of the microstructure of the deformed and annealed alloys was investigated. The alloys exhibit different hardness values for a given equivalent deformation. In industrial alloy, no recovery was observed at low temperatures. On the other hand, enhanced precipitation in this alloy leads to an increase in hardness balancing then softening due to recovery. A substantial decrease in hardness is observed around 250°C and seems to depend on alloy purity. © 2009 Elsevier B.V. Open access under [CC BY-NC-ND license](http://creativecommons.org/licenses/by-nc-nd/3.0/).

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Keywords: Aluminium alloys; ECAP; Recovery; X-ray diffraction; TEM;

1. Introduction

The reduction of grain size to the nanometer range has attracted much attention several years ago as it leads to produce ultra fine-grained materials exhibiting superior mechanical properties [1]. Therefore, several techniques are now available for this production, among others the most and attractive one is Equal-Channel Angular Pressing (ECAP) invented by Segal et al. [2, 3], it is now a widely-known procedure of refining grain size via severe plastic deformation down to submicron level [4, 5]. The ECAP technique can apply large cumulative strains to material without a change of the shape; through a specially designed die having two equally sized channels connected at a finite angle [6, 7].

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The aim of the present work is to characterize the change of the microstructure of an industrial and recycled Al-4%Cu alloys after equal channel angular pressing and subsequent heating and to prove that properties of recycled alloy can be improved which makes it competitive with the primary products after some heat and mechanical treatments. The stability of the microstructure during annealing is very important to preserve the enhanced mechanical properties. Transmission Electron Microscopy, X-ray diffraction analysis and Hardness measurements were used to characterise the evolution of the microstructure.

2. Experimental

An industrial 2017 aluminium-copper alloy (designed below IA) was received in form of rod (12x12x5000) mm³ in size. The recycled Al-4%Cu alloy (designed below RA) was received in form of casting ingots. Chemical compositions of the two materials are given in Table 1. We note that the whole compositions of the alloys are comparables excepting the presence of lead in the industrial alloy. Specimens of 10x10x50 mm³ were prepared and homogenized at 550°C/24hours and 500°C/6hours for IA and RA respectively. They were after quenched in icy water. ECA pressing was performed at room temperature using a die with two square channels forming an angle $\Phi = 90^\circ$; the angle Ψ representing the outer arc of curvature is 90° [8]. RA was pressed once (N=1) whereas IA was pressed successfully only after overaging for one hour at 350°C; in its quenched state, it breaks-down since the first passage. ECAP was performed at room temperature via route B (in which the sample was rotated by 90° around its longitudinal axis between successive passes). The die characteristics lead to an equivalent strain introduced equal to: $\epsilon_N = 0.906N$; N is the number of passes [8].

Table1: chemical composition of the investigated alloys (weight %)

Element (wt%)	Al	Mg	Si	Mn	Ni	Fe	Cu	Cr	Zn	Pb
RA	94.95	0.20	0.25	0.10	-	0.40	3.90	-	0.20	-
IA	92.80	0.30	0.70	0.95	< 0.1	0.65	4.45	0.05	< 0.15	0.10

The microstructures of the deformed specimens were investigated by X-ray peak profile analysis. X-ray diffraction patterns were recorded on a wide angle diffractometer in the θ - 2θ step scan mode by using CoK_α radiation. Scans were collected over a range of 20° - 120° in 2θ . To measure the intensity of each Bragg reflections an angle steps of $2\theta = 0.016^\circ$ and fixed counting times of 4s were taken. The crystallite size and the root mean square strain (rms-strain) were obtained from a deconvolution of peak profiles using the Halder-Wagner approach ; in that case, the reciprocal broadening $\beta^* = \beta \cos \theta / \lambda$ is related to the microstructural parameters by :

$$(\beta^*/d^*)^2 = D^{-1} \beta^*/(d^*)^2 + (\epsilon/2)^2 \quad (1)$$

where $d^* = 2 \sin \theta / \lambda$. According to equation (1), plotting $(\beta^*/d^*)^2$ versus $\beta^*/(d^*)^2$ gives the average values of coherency length D from the slope and of the rms-strain ϵ from the ordinate intercept. Specimens for TEM were thinned electrolytically using the twin jet technique in a mixture of 60% acetic anhydride and 40% of perchloric acid; the operating voltage was 15V.

3. Results and discussions

Figure 1 show the evolution of XRD pattern of the IA after extrusion and annealing at different temperatures. A careful examination reveals that the peaks relative to precipitates mainly Al_2Cu [9, 10], are broadened by extrusion, indicating a destruction of these precipitates by matrix dislocations. A substantial decrease of the peak breadth is observed by subsequent heating due to the gradual reduction of defects and to the grain relaxation. The same evolution was remarked in the recycled alloy (RA). Annealing at different temperatures of the RA leads to a decrease of the XRD peak breadth indicating that recovery takes place even at 100°C . Two stages can be remarked (figure 2): Below 200°C , the variation rate is low and can be attributed to dislocation rearrangement up to the cell formation. At elevated temperatures, recrystallization process is operative leading to a strong peak refinement. The same conclusion can be drawn from hardness measurements plotted on figure 3.

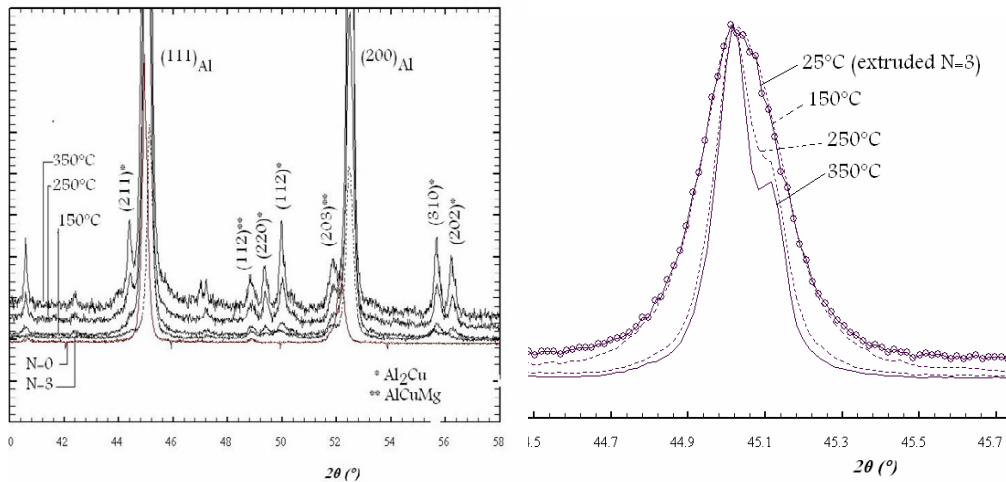


Fig. 1. (a) XRD patterns of IA extruded and heated one hour at different temperatures. (b) The evolution of (111) peak breadth as a function of annealing temperature.

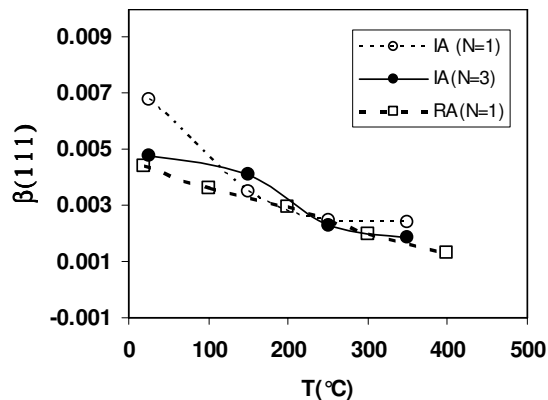


Fig. 2. Change of the integral breadth of the (111) peak in the two investigated alloys after ECA pressing and annealing for one hour at different temperatures.

The strong decrease of hardness relative to recrystallisation is observed at 200°C. In the industrial alloy (IA), annealing at 150°C leads to shifting of diffraction peaks due to a decrease of the lattice parameter of the matrix indicating a relaxation of the latter. However, a slight reduction of the peak breadth is detected at 150°C whereas this reduction becomes important at 250°C (figures 1, 3). Reduction of β was ascribed to the reduction of D and an increase of ϵ by severe plastic deformation. The values of D and ϵ obtained from the HW representations are plotted on figure 4. We note that heating above 250°C leads to a reduction of ϵ reaching then an equilibrium value close to zero. A substantial increase of D is also obtained but the final value at 350°C is still measurable and does not exceed 110 nm. The reduction of the lattice parameter may be due to the formation of dislocation cells leading also to a slight reduction of ϵ , and to a loss of the coherency between matrix and precipitates, which corresponds to the observed decrease of hardness and to the refinement of precipitate peaks after annealing (figures 3). It should be

noted here, that a reduction of copper concentration by precipitation leads rather to an increase of lattice parameter since it has a negative size effect in aluminium matrix.

TEM observations show that the RA alloy exhibits an heterogeneous microstructure after annealing at 200°C,: The pre-existing grains are splitted into subgrains of different sizes. Other regions show a more perfect grain boundaries indicating that the strong reduction of XRD peak breadth at this temperature (figure 2) is related to the formation of these dislocation cells. At a more elevated temperature (300°C), a substantial grain growth is observed (figure 5b).

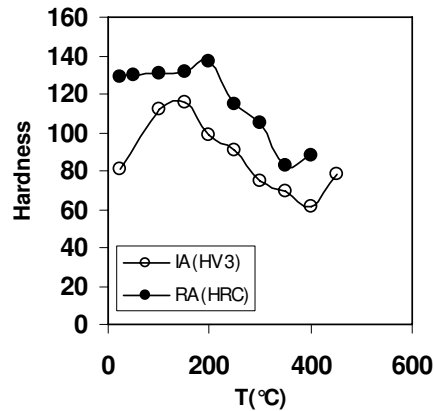


Fig. 3. Effects of annealing temperature on the hardness of the alloys of the present study extruded one passage.

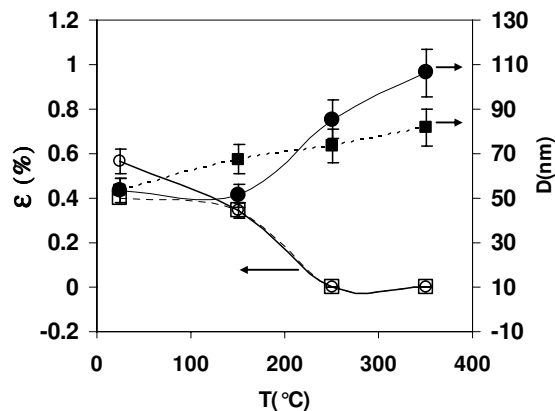
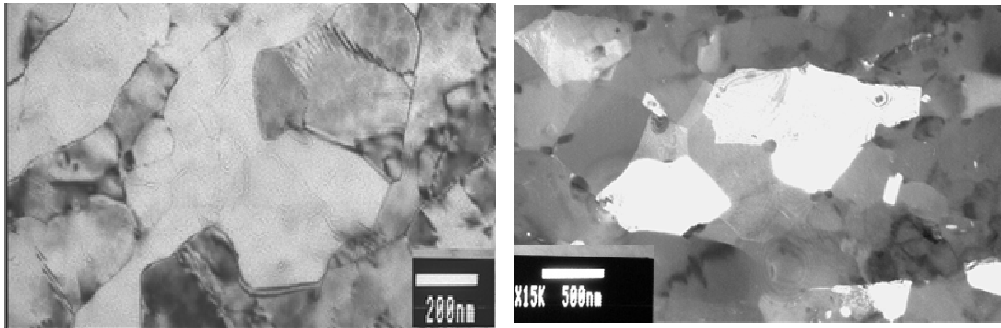


Fig. 4. Changes in the coherency length D (full marks) and in the rms- strain ϵ (open marks) as a function of annealing temperature for the ECA pressed material up to $N=1$ (squares) and $N=3$ (circles). Error bars represent calculation accuracy, which does not exceed 10% of the value.

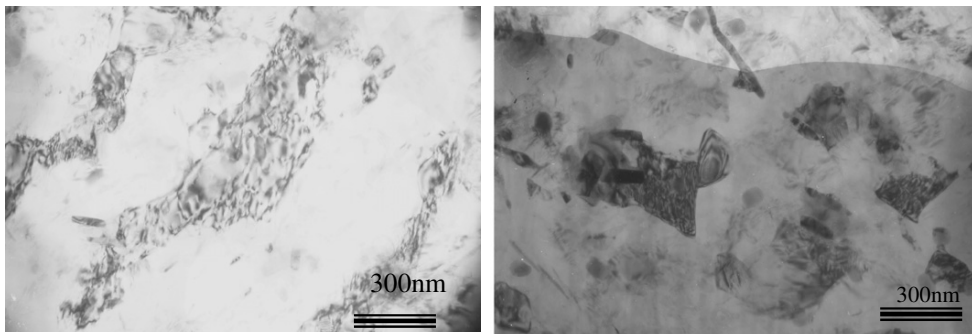
The stability of the microstructure with respect to grain growth depends on the presence of a second phase. In fact, the substantial growth observed at temperatures higher than 200°C is accompanied by a strong decrease of hardness. Hardness is related rather to precipitate spacing mainly if the latter is smaller than the mean grain diameter.



(a)

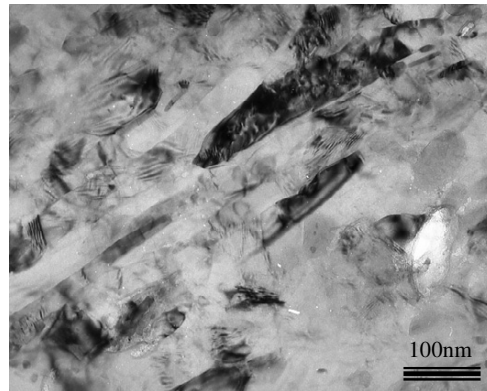
(b)

Fig. 5. Transmission electron micrographs of the recycled alloy (RA) extruded and annealed for one hour at 200°C (a) and 300°C (b).



(a)

(b)



(c)

Fig. 6. Transmission electron micrographs of the industrial alloy (IA) extruded (a) N=1, (b) N=2B, (c) N=3A.

The strong decrease could be related to the coarsening of precipitates (the transition from shearing to by-passing mechanisms of deformation), which reduces their dragging effect in recrystallization. On the other hand, it has been

shown that maintaining a small grain size needs not only fine precipitates but the latter must be stable with respect to coarsening [11].

Figure 6 shows a set of transmission electron micrographs of the industrial Al-4%Cu alloy processed by ECAP up to three passes. As it is mentioned above, the accumulation of this high amount of deformation was possible only after overaging. We note that during the first passage, extrusion leads to the accumulation of high dislocation density forming then dislocation cells (figure 6a). For subsequent passages, we note the formation of low – dimension grains close to 300nm in diameter indicating that grain growth takes place already during deformation due to heating of specimen. Grain size is however larger than that in the recycled alloy annealed at 200°C but the limited value of coherency length obtained after annealing, which is widely larger before extrusion reflects clearly the role of precipitates in dragging grain growth. The effect is however important mainly in the low-precipitate size range.

4. Conclusion

The high strength of heat treatable industrial Al-Cu alloys can be improved by severe plastic deformation. Among others, Equal Channel Angular Extrusion (or Pressing) is known to lead to a high strength via a reduction of grain size without a change of the specimen shape. The stability of fine grained materials with respect to subsequent heating is very sensitive to alloy purity and hence to elaboration procedures. The aim of the present work is to compare the behavior of an industrial alloy (IA) of a controlled purity with respect recovery and grain growth after severe plastic deformation to that of a recycled alloy (RA) of the same content of copper and to show that the latter can exhibit a comparative properties to those of the industrial alloy after controlled heat and mechanical treatment. Many conclusions may be drawn from this study:

- In both alloys, ECAP leads to a strong broadening of the XRD peaks due to a reduction of coherency length and an increase of the equivalent strain.
- Further annealing leads to a relaxation of the matrix, which seems to be slower in the industrial alloy.
- Transmission Electron Microscopy observations show a coarse grained structure in RA whereas coherency length in the IA is maintained low even at elevated temperatures. The main reason is that IA contains higher amount of elements than RA.

5. Acknowledgments

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